Reply to "Comment on 'Molybdenum sound velocity and shear modulus softening under shock compression'"

Jeffrey H. Nguyen,[1] Minta C. Akin,[1] Ricky Chau,[1] Dayne E. Fratanduono,[1] W. Patrick Ambrose,[1] Oleg V. Fat'yanov,[2] Paul D. Asimow,[2] and Neil C. Holmes[1]

[1]*Lawrence Livermore National Laboratory, Livermore, California 94550-9234, USA*

[2]*California Institute of Technology, Pasadena, California 91125, USA*

(Received 1 June 2015; published 27 July 2015)

We respond to the Comment by Errandonea *et al.* [Phys. Rev. B **92**, 026101 (2015)] on their reinterpretation of our published data [Nguyen *et al.*, Phys. Rev. B **89**, 174109 (2014)]. In the original paper, we argued that there is no solid-solid phase transition along the Hugoniot at 2.1 Mbars. There is, however, a softening of the shear modulus starting at 2.6 Mbars. Errandonea *et al.* [Phys. Rev. B **92**, 026101 (2015)] reinterpreted our data and concluded that there is a structural change near 2.3 Mbars on the Hugoniot. We will explore the differences and agreements in the two interpretations of our data.

DOI: 10.1103/PhysRevB.92.026102 PACS number(s): 64.70.K−, 61.50.Ks, 64.70.dj, 81.30.−t

We previously published molybdenum sound speed up to 4.4 Mbars along the Hugoniot [1]. Our main conclusions were that there is no statistically significant evidence for an abrupt phase transition at 2.1 Mbars as previously reported by Hixson *et al.* [2] and that there is evidence for shear modulus softening above 2.6 Mbars. We do not attribute any cause to such softening as there are multiple possible cases in which a material can soften, which our data cannot distinguish, including both mixed solid-liquid phase and loss of strength in a solid-only phase. Our results suggest that Mo remains in the bcc phase up to the melting pressure. We also cited independent diffraction data showing evidence of a likely bcc phase at up to 3.5 Mbars [3]. Errandonea *et al.* [4] reinterpreted our sound speed data and claimed that the abrupt change in shear modulus points to a structural change near 2.3 Mbars. There are significant differences and agreements in our interpretation and their reinterpretation. These points can be summarized in the following:

(1) We reported a change in the shear modulus, but our main focus was to determine if there was a discontinuity in the longitudinal sound velocity at 2.1 Mbars as reported by Hixson *et al.* [2]. Our statistical analysis, which used several fitting models and rejected less likely and more complex models, determined that Mo sound speed increases linearly up to 2.6 Mbars [1,5]. Our secondary conclusion was that there is a softening of the shear modulus at this pressure. We are pleased that Errandonea *et al.* [4] agree with our secondary conclusion; the difference is in the reported pressure. Errandonea *et al.* [4] calculated the shear modulus using our longitudinal sound speed data and Hixson's bulk sound speed data. From this, they concluded that there is an abrupt change in the pressure dependence of the shear modulus at 2.3 Mbars. Either the inconsistent use of the bulk sound speed data of Hixson *et al.* or the linear fit model selected by Errandonea *et al.* [4] may explain the difference in the calculated results and may have pushed the onset pressure of shear modulus softening lower.

(2) Errandonea *et al.* [4] maintain that an abrupt change in pressure dependence of the shear modulus can only be explained by a transformation from the bcc state to another structure. We do not believe a solid-solid or solid-liquid transition is necessary to explain the softening of the shear modulus. Copper exhibits similar softening at high pressure without undergoing a solid-solid phase transition [6]. Iron and tantalum exhibit similar softening, although only at pressures near their melting points [7,8]. There is no report of phase transition in these materials. Errandonea *et al.* [4] cited the work by Santamaria-Perez *et al.* [9] as reinforcing evidence that at pressures above 2.1–2.3 Mbars Mo may not be in a stable bcc phase. An extrapolation of their melt curve from 1.19 Mbars crosses the Mo Hugoniot near 2.1 Mbars. However, there are many calculations contradicting this conclusion [10,11], which both extrapolates well beyond the measured pressure range and requires assumptions about the nature of melt under pressure.

(3) We cited the work by Wang *et al.* [3] in our original paper confirming our interpretation. In their x-ray diffraction work, Wang *et al.* showed that there is no evidence for a solid-solid phase transition in Mo up to 9.0 Mbars on ramp compression. On shock loading, they can identify the (110) and (200) or (220) reflections of bcc Mo up to 3.5 Mbars. This experimental result disproves the argument of Errandonea *et al.* [4] for a phase transition at 2.3 Mbars. This work is under review for publication [12].

It is possible that a mixed bcc-liquid phase could explain both the observed diffraction lines and the decrease in shear modulus, however, so can a solid bcc phase that loses shear strength through other mechanisms. Without experimental evidence, which would require an extremely bright x-ray source capable of resolving liquid diffuse scattering mixed with a background and a solid diffraction pattern, these distinct cases cannot be distinguished.

In short, Errandonea *et al.* [4] agree overall with our reported softening of the shear modulus [1]. Our statistical analysis suggests that softening starts at a higher pressure than reported by Errandonea *et al.* [4], likely due to their use of the bulk sound speed data of Hixson *et al.* or to their model. We disagree with Errandonea *et al.* [4] as to whether our sound speed data require a structural transition. We do not have any evidence to make that conclusion. There are many explanations for shear modulus softening, including but not limited to partial melting. Recent diffraction work confirms our initial interpretation of a bcc phase of shocked Mo up to 3.5 Mbars [3,12].

Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC for the U.S. Department of Energy, National Nuclear Security Administration under Contract No. DE-AC52-07NA27344.

[1] J. H. Nguyen, M. C. Akin, R. Chau, D. E. Fratanduono, W. P. Ambrose, O. V. Fat'yanov, P. D. Asimow, and N. C. Holmes, Phys. Rev. B **89**, 174109 (2014).

[2] R. S. Hixson, D. A. Boness, J. W. Shaner, and J. A. Moriarty, Phys. Rev. Lett. **62**, 637 (1989).

[3] J. Wang, F. Coppari, R. Smith, J. Eggert, T. Boehley, G. W. Collins, and T. S. Duffy, *AGU Fall Meeting, San Francisco* (American Geophysical Union, Washington, DC, 2013), abstract no. MR23B-2356.

[4] D. Errandonea, R. Boehler, and M. Ross, Phys. Rev. B **92**, 026101 (2015).

[5] M. C. Akin and J. H. Nguyen, Rev. Sci. Instrum. **86**, 43903 (2015).

[6] D. Hayes, R. S. Hixon, and R. G. McQueen, in *Shock Compression of Condensed Matter*—1999, edited by M. D. Furnish, L. C. Chhabildas, and R. S. Hixon (American Institute of Physics, Melville, New York, 2000), p. 483.

[7] J. M. Brown and J. W. Shaner, in *Shock Waves in Condensed Matter*—1983, edited by J. R. Asay, R. A. Graham, and G. K. Straub (Elsevier, New York, 1984), p. 91.

[8] J. H. Nguyen and N. C. Holmes, Nature (London) **427**, 339 (2004).

[9] D. Santamaria-Pérez, M. Ross, D. Errandonea, G. D. Mukherjee, M. Mezouar, and R. Boehler, J. Chem. Phys. **130**, 124509 (2009).

[10] C. Cazorla, D. Alfè, and M. J. Gillan, Phys. Rev. B **85**, 064113 (2012).

[11] J. C. Cazorla, M. J. Gillan, S. Taioli, and D. Alfè, J. Chem. Phys. **126**, 194502 (2007).

[12] F. Coppari and T. S. Duffy (private communication).